Development of Yttrium Hydride Moderator for the Transformational Challenge Reactor[§]

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INTRODUCTION

Hydrogen is an outstanding moderator for neutrons of less than a few MeV of kinetic energies in nuclear systems by reason of its substantial equivalence in mass to the neutron, its low neutron absorption cross section, and its acceptably high neutron scattering cross section. Metal hydrides have been perceived as an efficient, low-risk option for high-density hydrogen storage since the late 1970s [1]. Transition metals are known to absorb large quantities of hydrogen to form metal-hydrogen solid solution at low hydrogen content and the hydride compounds at higher hydrogen content. The atomic density of hydrogen in many of these materials is far greater than in liquid hydrogen itself.

Metal hydrides are particularly well suited to thermal reactor system in which core weight and volume need to be minimized, where they serve as a constituent in fuels and in moderator and shield materials. Zirconium hydride (ZrH_x) and yttrium hydride (YH_x) are two most promising moderator materials in terms of the moderating ratio (the ratio of the macroscopic slowing-down power to the macroscopic cross section for neutron absorption). ZrH_x has been frequently used as a high-performance moderating material in advanced reactors. Examples include the Systems Nuclear Auxiliary Power (SNAP) Program [2]; Training, Research, Isotopes, General Atomic (TRIGA) research reactors [3]; and nuclear thermal propulsion reactor designs [4]. Large, hexagonal-cross-section rods of clad YH_x each with a central axial hole for a fuel element and coolant channel were moderator elements in the gas-cooled Aircraft Nuclear Propulsion reactor developed by General Electric [5]. Following several decades of hiatus in constructing advanced reactors, U.S. DOE recently launched a plan to build and operate an additively manufactured microreactor at Oak Ridge National Laboratory (ORNL), the Transformational Challenge Reactor (TCR), to demonstrate a faster, more affordable approach to advanced nuclear energy [6]. YH_x was selected as the leading moderator material for this reactor [7]. Therefore, there is a strong need for thermally and chemically stable massive metal hydrides for use in shield, reflector, moderator, and moderator-fuel in nuclear reactor components.

CHALLENGES TO FABRICATE CRACK-FREE YHX

The common method of preparing metal hydrides is by direct reaction of the metal with hydrogen, guided by the phase diagram and the pressure-composition-temperature relationship. The reaction of hydrogen with the metals (e.g, Zr, Y, Ti, Th) is a diffusion-controlled exothermic process that normally results in expansion of the metal lattice as the hydrogen enters portions of the lattice. This gives rise to a substantial decrease in density and represents a significant volume expansion during the hydriding process (e.g., 19% in the case of TiH_2, 17% in the case of ZrH_2, 6% in the case of YH_2). Such changes produce severe stresses in the massive metal hydrides since the diffusive nature of the hydriding process results in a large hydrogen concentration gradient. As shown in Fig. 1 (a), during the hydriding process, a hydride case forms on the surface and hydrogen is in solid solution in the interior of the specimen. At the interface between the hydride case and the interior, the growth accompanying hydride formation generates stresses that can easily exceed the fracture strength of the hydride and cause extensive cracking. A cracked as-fabricated YHx was shown in Fig. 1(b). Therefore, to prepare massive single-piece forms of metal hydride forms, this cracking must be avoided—which is challenging.

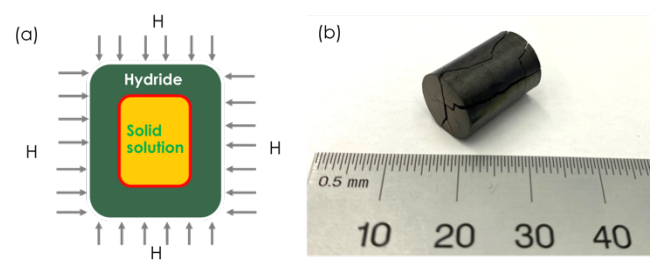

Fig. 1. (a) Schematic plot of the hydriding process; (b) a cracked as-fabricated YH_x rod.

HYDRIDING PROCESS

Yttrium hydrides with desired H/Y atom ratios may be fabricated in a variety of ways, e.g., by introducing

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measured amount of hydrogen into small-volume retorts or by carefully matching retort temperature and hydrogen pressure. In the current work, the first method was employed. Yttrium hydride may also be fabricated via the powder sintering route as demonstrated by researchers at Los Alamos National Laboratory [8]. However, the sintering process is cumbersome, allows for far simpler geometries than what is viable by bulk metal hydriding, and is likely to result in introduction of impurities in the hydride. Given these issues and no obvious benefit, the sintering route for yttrium hydride processing will not be pursued under the TCR program.

A hydriding system (Fig. 2) with an ultra-high vacuum level of 1×10^{-7} torr was established at the ORNL Low Activation Materials Development and Analysis lab for the fabrication of yttrium hydride. The maximum temperature of the furnace in this system is 1100°C. A quartz retort was used to minimize the permeation of hydrogen at elevated temperatures.

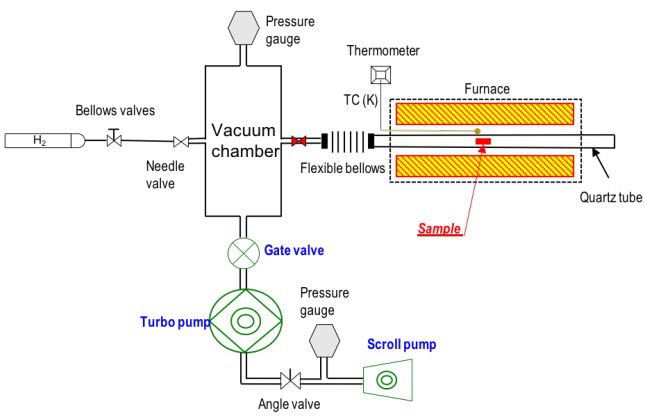

Fig. 2. Schematic plot of the static hydriding system.

RESULTS

Following the above-mentioned fabrication procedure, we have successfully fabricated crack-free YH_x disks, pellets, and hexagonal coupons with complicated geometries, as shown in Fig.3.

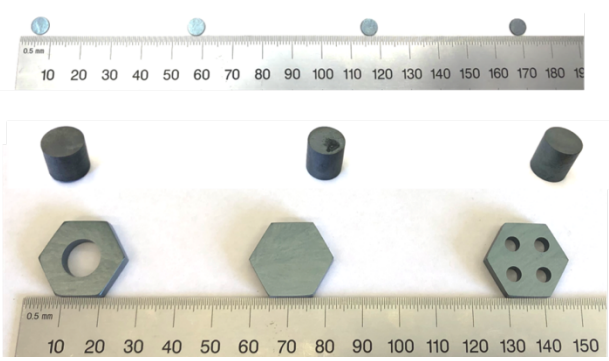

Fig. 3. As-fabricated yttrium hydride disks, pellets, and hexagonal coupons.

The hydrogen content of the as-fabricated YH_x samples was first determined based on the weight change, which was assumed to be due to the absorption of hydrogen. Hot vacuum extraction and LECO inert gas fusion techniques were also used to measure the hydrogen concentration of the disks sectioned from the as-fabricated rods. Table 1 list the results of 5 YH_x pellets with 10mm in diameter and 10mm in height. It is apparent that the H/Y atom ratio determined by weight change method and the vacuum hot extraction technique are comparable, within 4% uncertainty, expect YHR-3. LECO measurements significantly underestimate the hydrogen concentration due to the fact that the released H from the sample is close to the detecting limit of the system. We will use the H/Y atom ratio determined by the weight change method in the following context.

TABLE 1. Hydrogen concentration of the as-fabricated YH_x pellets

Sample	H/Y ratio determined using weight change	Vacuum hot extraction	LECO
YHR-1	1.70	1.72	1.45
YHR-2	1.52	1.57	1.32
YHR-3	1.59	1.71	1.37
YHR-4	1.84	1.89	1.58
YHR-5	1.88	1.87	1.61

No surface cracking was observed on the as-fabricated YH_x pellets, disks, or hexagonal coupons. X-ray computed tomography (XCT) analysis was used to explore possible internal cracking. XCT measurements were conducted using a ZEISS Metrotom M800 (200 kV/14 W power). A single scan (1 hour) was conducted to evaluate the whole volume of the specimen. The resolution varies from 9~13 voxel size. Figure 4 shows the XCT images of two YH_x pellets, i.e., YHR-1 ($YH_{1.70}$) and YHR-5 ($YH_{1.88}$), as examples. No cracking was found in any of the tested YH_x pellets (YHR-1, YHR-2, YHR-3, YHR-4, and YHR-5).

X-ray diffraction analysis was employed to confirm the phases present in the fabricated hydride samples. XRD samples were prepared by depositing YH_x powder on a low-background silicon single-crystal sample holder. Samples were also mixed with lanthanum hexaboride (LaB_6) powder, used as an internal standard during pattern refinement. High-resolution diffraction patterns were obtained using a Bruker D2 Phaser benchtop X-ray diffractometer of 0.30 kW with Cu Kα radiation. Rietveld refinement was performed on the experimental patterns using General Structure Analysis System [9]. Figure 5 shows an example of the powder XRD pattern of YHR-5 ($YH_{1.88}$). Of the sample, 99wt% presented a cubic YH_2 phase and the rest was α-yttrium. Table 2 lists the XRD results of the 5 yttrium hydride pellets. Note that all fabricated YH_x samples exhibited two phases following the current hydriding routine. The fraction of the YH_2 phase generally increases with the increasing hydrogen

concentration, although YHR-2 and YHR-3 don't follow this trend. YHR-4 and YHR-5 exhibit the nearly-single phase yttrium hydride. The presence of α-yttrium in these samples can likely be ascribed to the rapid hydrogen desorption on the sample surface at high temperature during the pumping process prior to bringing down the system temperature to room temperature. The absence of any noticeable oxide phase underlines the well-controlled vacuum environment within the system.

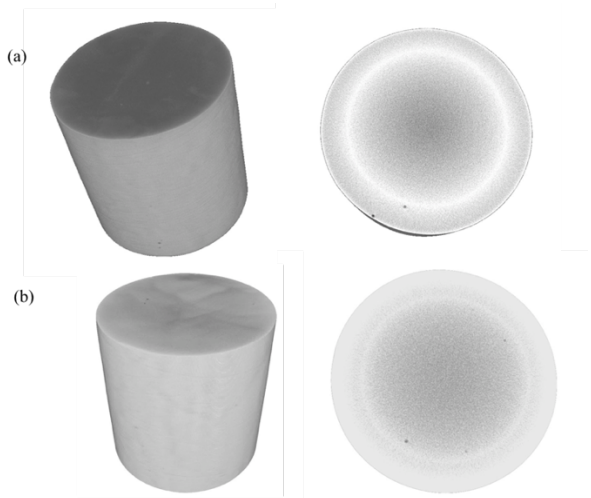

Fig. 4. XCT images of YH_x pellets (a. YHR-1 and b. YHR-2) and the snapshots of cross sections.

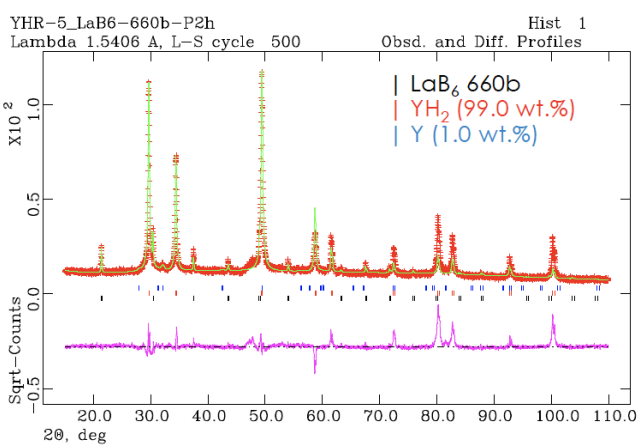

Fig. 5. XRD pattern of $YH_{1.88}$ along with Rietveld refinement fit.

SUMMARY

This report provides a summary of efforts to fabricate the crack-free bulk yttrium hydride to support thermal spectrum design options for the Transformational Challenge Reactor (TCR). The challenges to fabricating massive, crack-free single-phase yttrium hydride (YH_x) are discussed. By using a modified static hydriding system, we have successfully demonstrated the fabrication of crack-free yttrium hydride, guided by the well-established thermodynamics of the binary yttrium-hydrogen system. Hydrogen concentrations in as-fabricated YHx samples were determined by employing vacuum hot extraction technique, LECO inert gas fusion technique, and weight change. X-ray diffraction was used to identify the present phases within the YH_x samples. X-ray computed tomography measurement confirmed the products are all crack-free. More efforts are underway to characterize the as-fabricated yttrium hydride samples and to improve the quality of the products by reducing the metal yttrium fraction.

TABLE 2. XRD results of as-fabricated YHx pellets

Sample	H/Y ratio	XRD	
		YH_2 phase	Y phase
YHR-1	1.70	81%	19%
YHR-2	1.52	77%	23%
YHR-3	1.59	72%	28%
YHR-4	1.84	98.9%	1.1%
YHR-5	1.88	99%	1%

ACKNOWLEDGEMENTS

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REFERENCES

1. Sakintuna, B., F. Lamaridarkrim, and M. Hirscher, *Metal hydride materials for solid hydrogen storage: A review*☆. International Journal of Hydrogen Energy, 2007. **32**(9): p. 1121-1140.
2. Davies, N. and R. Forrester, *Effects of irradiation on hydrided zirconium-uranium alloy NAA 120-4 experiment*. 1970, Atomics International Div.: Canoga Park, CA.
3. Simnad, M., *The U-ZrHx alloy: Its properties and use in TRIGA fuel.* Nuclear Engineering and Design, 1981. **64**(3): p. 403-422.
4. Haslett, R., *Space Nuclear Thermal Propulsion Program*. 1995, Grumman Aerospace Corp.: Bethpage, NY.
5. *Summary Report of HTRE No.3 Nuclear Excursion.* GE APEX 509, 1959.
6. https://tcr.ornl.gov.
7. B. Ade, A.W., B. Betzler, M. Greenwood, P. Chesser, P. Jain, J. Burns, K. Terrani, J. Rader, B. Hiscox, J. Heineman, J. Risner, K. Smith, F. Heidet, A. Bergeron, J. Sterbentz, T. Holschuh, N. Brown, , *Candidate Core Designs for the Transformational Challenge Reactor.* Proc. PHYSOR 2020, Cambridge, UK, 2020.
8. Shivprasad, A.P., et al., *Development of sintered yttrium dihydride compacts for nuclear reactor*

moderator applications. Nuclear and Emerging Technology for Space, American Nuclear Society Topical Meeting, 2019.

9. Larson, A.C. and R.B. Von Dreele, *General Structure Analysis System* Los Alamos National Laboratory Report LAUR 86-748, 2004.